



Modeling redistribution of α -HCH in Chinese soil induced by environment factors

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ABSTRACT

This study explores long-term environmental fate of α -HCH in China from 1952 to 2007 using ChnGPERM (Chinese Gridded Pesticide Emission and Residue Model). The model captures well the temporal and spatial variations of α -HCH concentration in Chinese soils by comparing with a number of measured data across China in different periods. The results demonstrate α -HCH grasshopping effect in Eastern China and reveal several important features of the chemical in Northeast and Southeast China. It is found that Northeast China is a prominent sink region of α -HCH emitted from Chinese sources and α -HCH contamination in Southwest China is largely attributed to foreign sources. Southeast China is shown to be a major source contributing to α -HCH contamination in Northeast China, incurred by several environmental factors including temperature, soil organic carbon content, wind field and precipitation.

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1. Introduction

Persistent organic pollutants (POPs) are characterized by their environmental persistence, toxicity, potential for bioaccumulation, and potential for long-range transport (Beyer et al., 2000; El-Shahawi et al., 2010). After entering into environment, POPs can be redistributed globally by the effect termed “global distillation” (Wania and Mackay, 1993). Long-range atmospheric transport (LRAT) and subsequent atmospheric deposition have been recognized as the most important pathways of POPs to migrate from their places of origin to remote pristine areas (Cheng et al., 2007). POPs are volatilized from warmer region and delivered to north through several cycles of deposition and reemission before reaching their final destination, a process termed “grasshopping” (Wania and Mackay, 1996). Once reaching the Arctic, they become more persistent due to slower removal through biodegradation and atmospheric reactions (Hung et al., 2010). Enrichment of POPs in the Arctic and remote northern territories suggests that such latitudinal concentration effect of POPs would also take place on a regional scale. Knowledge in this aspect is very important to regulate organic chemicals and assess ecosystem risk and human exposure in the region (Prevedouros et al., 2004).

Environmental distribution of POPs depends partly on their primary and secondary emissions in the environment (Jia et al., 2010; Li et al., 2010). The primary emission of a chemical is characterized by application intensity and the period of time of its use, and processes entering into the environment (Li et al., 1998b). The secondary emission always links with redistribution of a chemical through interactions between its physicochemical properties and environmental factors (Wania and Mackay, 1996). In the present study, we shall refer these environmental factors as wind field, ambient temperature, precipitation, organic matter content of soil, etc (MacLeod et al., 2001, 2005; Hansen et al., 2004; Ma et al., 2005a). Given that the interactions and relationships among POPs and those environmental factors are complex on a regional scale (Ma et al., 2005b; Hansen et al., 2006; Sun et al., 2006), they are not straightforward to be understood from measurement data (Suzuki et al., 2004). To elucidate these complex relationships, multimedia mass balance models have been developed and extensively used to interpret the redistribution of POPs in multi-compartments induced by LRAT and re-volatilization (MacLeod et al., 2001, 2005, 2010; Prevedouros et al., 2004). Redistribution process of a chemical under the impact of environmental factors is a slowly varying process as compared with that induced by fresh usage or emission of the chemical (Li et al., 1998b; Li and Macdonald, 2005). To better identify redistribution pattern of a chemical, one has to take the long-term effect of environmental factors into consideration in a modeling exercise. Spatial pattern of POPs in soils often exhibits

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stronger signatures of the long-term effect of environmental factors on POPs distribution as compared with that in air because soil is a non-floating (or low floating) environmental matrix and can absorb abundant POPs with ample organic matter (Mackay et al., 2006). Therefore, soil is a better environmental matrix to explore the environmental behavior of a chemical under influence of long-term environmental factors.

Pesticides are mostly applied to crops and soils for agricultural purposes which can be directly (via spraying to air) and indirectly (via reemission from soils) emitted to the atmosphere (Li et al., 1998b). In this sense, the processes of pesticides emitted into the environment are more facile to be evaluated than industrial POPs (Li et al., 1998b; Breivik et al., 2002). α -HCH, an organochlorine pesticide, as the major isomer of technical HCH, was extensively used in the world (Li, 1999). It has been recently listed as one of nine new POPs for inclusion in the Stockholm Convention (www.pops.int).

China began to produce technical HCH in 1952, and this production continued until it was banned in 1983 (Li et al., 1998b). Total usage of this pesticide in China was approximately half of global total consumption (Li and Macdonald, 2005). Technical HCH was heavily used in relatively warmer Southern and Southeastern China (Li et al., 1998b, 2001). To date, the use of technical HCH in China has been banned for more than two decades (Li et al., 1998b, 2001). This enables us to investigate the influence of environmental factors, by excluding the primary emission, on the fate and redistribution of the chemical. Due to its stronger potential of LRAT, α -HCH from external (foreign) sources is expected to be transported and deposited into Chinese environment and such long-range transport of α -HCH to China likely ranges from continental to global scales. However, since China used the highest amount of technical HCH/ α -HCH in the world (Li and Macdonald, 2005), the external sources of α -HCH are assumed to be minor in comparison with the Chinese domestic sources, and identification of the contribution of α -HCH from internal sources to the air concentrations over China is essential for understanding and differentiating the relative importance of multiscale atmospheric transport to China. The present study aims at simulating long-term fate of α -HCH from 1952, when the pesticide began to use in China, to 2007 using the Chinese Gridded Pesticide Emission and Residue Model (ChnGPERM), in order to identify major sink regions of the chemical associated with major environmental factors in China. The main objectives of the present study are (1) to assess the performance of ChnGPERM; (2) to distinguish sink and source regions of α -HCH over China; (3) to explore the source–receptor relationship of α -HCH in Chinese soils.

2. Model and model input

ChnGPERM employed in this investigation is developed based on Gridded Basin-based Pesticide Mass Balance Model (GB-PMBM), which has been applied previously to assess α -HCH budget in the Taihu region, China (Tian et al., 2009a). Briefly, GB-PMBM is a gridded mass balance model with $1/6^\circ$ latitude by $1/4^\circ$ longitude resolution. The model consists of transfer and transport modules. The transfer module, using a level IV fugacity method, describes the changes in pesticide concentrations and inter-compartmental transfer of the substance in the multimedia environment, including 5 soil types in 4 soil levels, water, sediment, and the air compartment in the surface boundary-layer. The transport module simulates mass exchange of the substance between model grid cells driven by atmospheric transport and water current.

In the present study, the model domain covers entire China spanning from 17° to 55°N and from 70° to 135°E . A grid system with the same horizontal resolution as that for GB-PMBM has been

established over the model domain, as shown in Fig. S1 of Supplementary Information (SI). The environmental processes and numerical method adopted in the transfer module are also taken the same as that used in GB-PMBM (Tian et al., 2009a). Whereas, as compared with the transport module used in GB-PMBM, transport by water current is neglected because the most underlying surfaces over the model domain are covered by lands. The details are described in SI.

By assuming 67.5% composition of α -HCH in technical HCH (Li et al., 1998b), gridded annual usage of α -HCH from 1952 to 1984 is created using technical HCH usage inventories with the same resolution of $1/6^\circ \times 1/4^\circ$ latitude/longitude in China (Li et al., 2001). The processes of α -HCH entering into environment are evaluated by three application modes and frost-free zones in China. The model inputs include gridded daily meteorological data, soil characteristic parameters, and surface features, etc. The details are also presented in SI.

3. Model evaluation

In the Chinese POPs Soil and Air Monitoring Program (SAMP-I) operated by the International Joint Research Center for Persistent Toxic Pollutants (IJRC-PTS), 162 soil samples across China have been collected during June 2005–June 2006 and analyzed in the laboratories of IJRC-PTS (Ren et al., 2007; Zhang et al., 2008). To evaluate the model performance, the modeled mean concentrations in the third soil layer (from 1.1 to 21.1 cm) with 5 soil types in 2005 have been compared with the measured soil concentrations of α -HCH. Fig. S10a and b in SI show the measured and modeled α -HCH soil concentrations, respectively. In general, the similar spatial pattern between the modeled and sampled concentrations in soil can be identified. A further comparison is made by calculating Spearman correlations between the measured and modeled soil concentrations at 123 model grids collected from the 162 soil sampling sites across China. If several sampling sites are located in one grid cell, an arithmetical mean value of the measured soil concentrations averaged over the grid cell is taken in the correlation analysis. Spearman correlation at 0.37 ($p < 0.001$) and the mean ratio of 0.7 between measured and modeled soil concentrations (S5.1 of SI) suggest that the modeled results agree reasonably well with the measurements across Chinese soils.

Further insight into the model performance can be gained by comparing modeled soil concentrations with historical measurements of α -HCH in Chinese soils, collected from literatures. Detailed information and results for this comparison are described in SI and Table S7 and Fig S12 of SI. The result shows that the model captured well the temporal and spatial variations of α -HCH concentration in soil with Spearman correlation coefficient at 0.46 ($p < 0.01$) and the median of 1.1 of the averaged ratio between the modeled and monitoring dataset.

In addition, overall mass balance of α -HCH in the model domain from 1952 to 2007 is estimated to assess the mass conservation of the model. The smaller relative error ($<0.01\%$) between total input and output mass proves that the model achieves very well the mass conservation. The details are shown in the subsection S5.3 and Fig. S13 of SI.

4. Results and discussion

4.1. Identification of sink region

Fig. S14 (a–f) of SI shows the annual average of modeled daily α -HCH soil concentrations in 1960, 1980, 1990, 2000, 2007, and 2012, respectively. The concentrations in 2012 are derived by using arithmetic mean of the daily gridded meteorological data from

2000 to 2007 in the model simulations. A glance to the annual variation of soil concentrations indicates appreciable increasing in soil concentrations of α -HCH in the northern part of Eastern China, especially since the ban of the pesticide after 1984. The figure also shows that the primary source regions, depending on the use pattern of the chemical, are characterized by high concentrations. Whereas, the secondary source regions exhibit more uniform concentration pattern which associate largely with soil organic carbon (SOC) and temperature, namely, the environment factors. These changes characterize the transition of α -HCH emissions from the primary source pattern to the secondary source pattern (Li et al., 2010).

Such changes can be further demonstrated by Pearson correlations between the soil burden of the pesticide and two environmental factors, SOC and temperature, during 1952–2007. Fig. S15 of SI displays the relationship between soil burden of the pesticide and annual mean environmental temperature (Fig. S15a), and soil burden and SOC (Fig. S15b). These correlations are computed within the region bounded by two pink lines in Fig. S1 of SI, passing through model grids between $X = 140$ and 200 and covering Eastern China with an interval of every 5 model grids. As shown, from the 1950s to the early 1980s during which the pesticide was applied in China, increasing positive correlation coefficients between soil burden and ambient temperature extend toward the east. This suggests that the greater abundance of α -HCH was entering soils in south with higher temperature than in north with lower temperature, and this difference increased from west to east. This spatial pattern of the soil burden was determined by the primary emission. After 1984, when α -HCH was banned and primary emissions were ceased in China, air temperature played an increasing role in the redistribution of α -HCH. Higher air temperature favors stronger re-volatilization of the pesticide from soils, leading to a decrease in their soil burden, in particular in those primary source regions. This would yield negative correlations between air temperature and soil burden, as seen from Fig. S15a of SI after 1995. The spatial distribution of soil burden starts to exhibit the secondary source pattern. Similar transition of positive–negative correlations associated with SOC can be found in Fig. S15b of SI. The SOC in the soils of Southeastern China subject to higher technical HCH usage is lower than that in Northern China subject to lower technical HCH usage (see Fig. S7 of SI). This causes negative correlations between the SOC and soil burdens of α -HCH during 1952–1984. After 1984 when the application of α -HCH was ceased in China, a negative to positive inversion of the correlations occurred and the positive correlation increased continually with time (see Fig. S15b of SI). This turns out that the soils with greater SOC in Northeast China tend to absorb more abundant of the chemical than that with lower SOC in Southeast China, and hence exhibits more signatures of the secondary source pattern. SOC is often found to be positively correlated with concentrations of POPs in soil. For example, it was found that Σ HCH and Σ DDT in surface soils from Haihe plain, China depended significantly on SOC (Tao et al., 2008). The results elaborated above not only illustrate the important role of ambient temperature and SOC in the redistribution of α -HCH in soils, but also provide a good example for chemical grasshopping effect (Wania and Mackay, 1996) in Eastern China.

Annual change in modeled α -HCH soil burden (Fig. S14 of SI) also suggests Northeastern China to be a sink region of the pesticide. The achievement of mass conservation of the model enables us to distinguish sink regions of α -HCH in China. By assuming the gridded usage from 1952 to 1984 as the inputs, and the mass loss due to degradation in all compartments, leaching in fourth soil layers (>51.1 cm) from 1952 to 2007, and the residue in all matrixes in 2007 as the outputs, we can identify the sinks of α -HCH across China. Fig. S16 of SI presents a schematic view for the input–output mass balance at a model grid. The mass conservation requires that

amounts of the input and output mass at a grid cell should be equal if the mass exchange among the grid cell due to atmospheric transport is excluded. One can then compare the ratios of the output and input mass to evaluate the mass exchange among the grid cell due to atmospheric transport. The ratio at a grid cell equal to 1 indicates an equilibrium condition caused by atmospheric transport and deposition. The ratio >1 represents net input, suggesting the net inflow to the grid cell due to atmospheric transport and deposition. The ratio <1 indicates the net outflow from the grid cell. Fig. 1 illustrates the output/input mass ratios across China. It is noted that these ratios are computed only at those model grid cells with historical usage of the pesticide. As seen from Fig. 1a, the regions with the ratio >1 mainly cover the northeastern and central parts of China whereas the regions with the ratio <1 span from Southern to Southeastern China with lower values of less than 0.5 in the downstream regions of Yangtze River in Eastern China, where technical HCH was heavily used as shown in Fig. S4 of SI. Soil concentrations of α -HCH over this part of China are also shown at a lower level in 2005 which has been validated by the measurement as described in section of Model evaluation. The model results indicate that ~ 30 – 50% of α -HCH used previously in these source regions has been emitted, delivered, and deposited to those regions characterized by the ratio >1 within China and also to the regions outside China.

Further insight into the features of the mass exchange due to atmospheric transport can be gained by comparing the differences between the input and output masses, as shown in Fig. 1b. In the

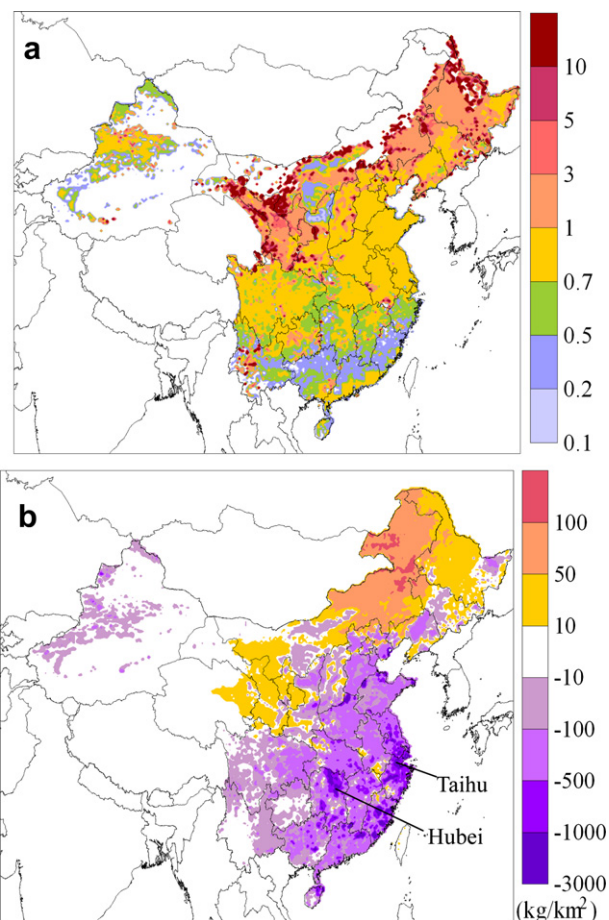


Fig. 1. Comparison between output and input of α -HCH from 1952 to 2007 (a: ratio (output/input), b: difference (output–input)).

figure, positive values, indicating net input, can be found in the northeast and central north regions of China. Interestingly, the stronger net input defined by greater positive values appeared in the western part of Northeastern China where technical HCH was never used. Except for Northeastern China, the rest of Eastern China appears to lose the mass of the chemical, characterized by negative values showing net output. The prominent output features can be found in Taihu region and the north of Hubei province where the pesticide had been heavily used, as shown in Fig. S4 of SI. This result supports further the analysis presented in Fig. 1a, suggesting that Northeastern China is a major sink of the substance over China.

4.2. Identification of source region

To identify quantitatively the relative contribution of different source regions of α -HCH in China to its budget in Chinese soil, the numerical simulations have been performed for three model scenarios. These three scenarios consist of (1) Northern China sources only (scenario 1), (2) Central China sources only (scenario 2), and (3) Southern China sources only (scenario 3). Fig. S17 of SI displays the three regions and corresponding total gridded historical usages of α -HCH in each of the regions. The contribution of the α -HCH emissions from each source region to receptors across China are assessed through calculating ratios of the mass of α -HCH soil residual from each individual scenario to the total mass summed from the model scenarios 1–3, defined by

$$r_{si} = M_{si} / \sum_{i=1}^3 M_{si} \quad (1)$$

where r is the ratio of soil residual mass (M) of α -HCH at a model grid cell produced from model scenarios 1–3 (subscript $si = 1, 2, 3$). Since the governing equations, including the transfer and transport equations, in the model are linear, the solutions can be linearly combined, and hence the above approach can be used to determine the contributions from individual α -HCH source region to receptor regions.

Fig. S18(a–c) of SI illustrates the spatial pattern of the computed ratios for three model scenarios in 2000. In general, local sources make the largest contribution to α -HCH contamination over their nearby regions due to source proximity. Nevertheless, it can be also seen from Fig. S18c of SI that the Southern China emissions (scenario 3) make a significant contribution at 80–90% to α -HCH contamination over Inner-Mongolia and the west part of Northeastern China where no usage of the pesticide was ever reported. The contribution from Southern China (scenario 3) is even higher than that from Central (scenario 2) and Northern China (scenario 1), suggesting that Southern China seem to be a significant source region to Inner-Mongolia and the west part of Northeastern China, due to stronger application intensity of α -HCH and warmer conditions in Southern China which result subsequently in much stronger emissions.

Another interesting finding is the source–receptor relationship between the three highlighted sources in the three model scenarios (Fig. S17 of SI) and Southwestern China, including Tibetan Plateau on the west of purple line of Fig. S18 of SI. Though heavily used in some areas of Southeastern China, the local input of the pesticide from Southeastern China into Tibetan Plateau appears less significant. This source region makes only 10–20% contribution to the eastern edge of the Tibetan Plateau, as shown in Fig. S18c of SI. The reason is that α -HCH – laden air mass from Southeastern China sources seldom approached the plateau under prevailing westerly wind over mid-latitudes (Bey et al., 2001; Liu et al., 2003) and increasing terrain height toward the plateau (Shen et al., 2009). Northern China source contributed to 80–90% soil burdens of α -HCH in the plateau and Southern China source was the largest

contributor at the region near the purple line of Fig. S18 of SI. This implies that the sources in the west part (Xinjiang province, see Fig. S17 of SI) of North China make a significant contribution to α -HCH soil burdens in the plateau and Southwestern China. This can be demonstrated by mean winds at 10 m height in spring–summer seasons and autumn–winter seasons during 1980–2000, as shown in Fig. S19 of SI. The mean vector winds in spring–summer seasons, when warmer conditions favor stronger re-volatilization, exhibit a clear atmospheric route from Northwestern China (Xinjiang province) to the plateau. Given that emission sources of the chemical outside China (or external sources) is not taken into account in the aforementioned modeling scenarios, these results suggest that the soil residues of α -HCH in the plateau is mostly attributable to the emissions from Northwestern China source region.

4.3. External versus internal sources

Although the present study has been focusing on α -HCH contamination to Chinese soils from the emission sources within China, a concern might be raised for the potential impact of external sources (outside China) on the soil burdens of the chemical across China. In particular, technical HCH had been still used in India till 1990 after its ban in China in 1984 (Li, 1999). In a previous study, we have investigated potential influence of India and Europe/Russia emissions of γ -HCH on its atmospheric level and deposition to China (Tian et al., 2009c). Results showed that these external emission sources contributed almost entirely to γ -HCH contamination to Western China only and appreciable atmospheric level of the chemical derived from these external sources was observed only over the Tibetan Plateau and part of Southwestern China, originated from the India emissions. As a result, α -HCH contamination in Southwest China would be also likely attributed to foreign sources, as illustrated by Fig. S10 of SI, which shows higher soil concentration measured in the plateau as compared to the modeled concentration accounting for China sources only.

To elucidate the potential influence of external α -HCH emissions on the soil burdens in China and examine the reliability of our modeling strategy, an extra model scenario was designated by accounting for external emissions in the ChnGPERM. Fig. S20a of SI shows accumulated α -HCH usage in China from 1952 to 1984 (same as Fig. S4 of SI) and in the regions outside China from 1952 to 1997 (Li, 1999). Among these regions, India is the second largest user of the chemical next to China. By implementing the updated emissions that take both external and internal sources into consideration, the ChnGPERM was integrated from 1952 to 2005 and resulted annual soil concentrations in 2005 were compared with that derived from internal (China) emissions. Fig. S20b of SI presents the ratios (Eq. (1)) of the soil concentrations computed using China emissions (Fig. S4 of SI) to that derived from the external + internal (China) emissions (Fig. S20a of SI). As shown, over 90% of α -HCH soil burdens in Eastern China and the part of Northwestern China (Xinjiang province) is attributed to the China emissions. The regions between Eastern China and Northwestern China, including the Tibetan Plateau, however, appear to be contaminated mostly by external emission sources, notably the sources in India and other Southern Asian countries. Given that our interest in the present study is mostly on eastern part of China where the external sources exert a weak influence only (also see Figs. 17 and 18 of SI), as will be elaborated below, their roles in source–receptor relationships will not be pursued further.

4.4. Source–sink relationship

We further explore temporal trends of the contributions from the three sources highlighted in Section 4.2 to four regions in

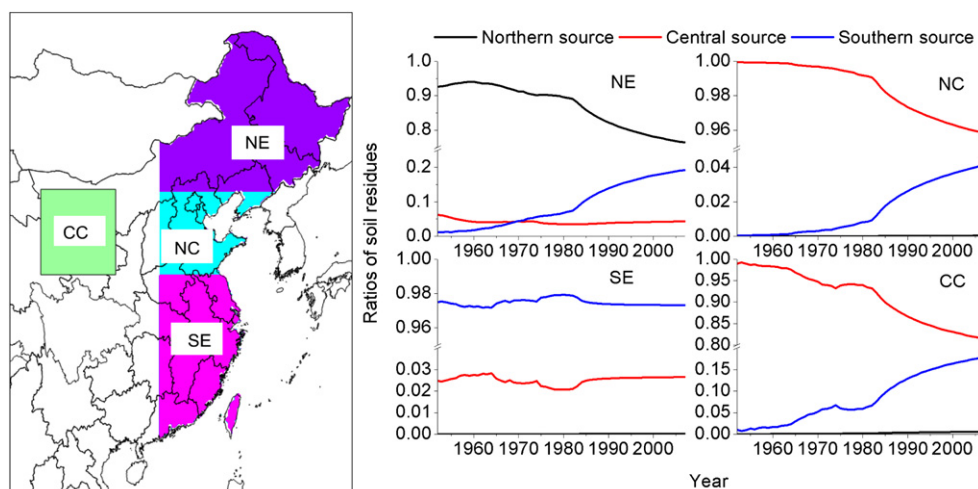


Fig. 2. Four receptor regions specified to identify the contribution ratios from the three model scenarios (left panel) and the annual change in the ratios from 1952 to 2007 (right panel).

Eastern and Central China. These four regions are termed Northeast (NE), North China (NC), Southeast (SE), and Central China (CC) regions across Eastern and Central China, respectively, as shown in Fig. 2 (left panel). Residual burdens of α -HCH in soils in the four regions simulated from each of the model scenarios 1–3 (Section 4.2) are summarized and their contribution ratios are assessed by Eq. (1) for the period of 1952–2007. The trend of the ratios is also displayed in Fig. 2 (right panel). The soil burdens in the selected four regions are affected primarily by corresponding local usage over the whole period of time. As seen from Fig. 2, the soil burdens in NC and SE are mostly contributed from the Central and Southern China sources, respectively, whereas the contribution from the Northern China source is not significant. Relative contributions to the selected receptor regions from the three source regions (Fig. S17 of SI), however, alter with time. It was shown in Fig. 2 that the contribution from Northern China sources to the soil burden in NE region reached the highest at over 90% in the 1960s and declined thereafter down to below 80% in the 2000s, whereas the Southern China sources made an increasing contribution to the α -HCH soil burden in this region during the same period. Likewise, almost all soil burdens in NC and CC receptor regions could be attributed to the input of α -HCH from the Central China sources in the 1950s and 1960s but such the input decreases substantially after the 1980s. Rather, the Southern China emissions become increasingly importance to the α -HCH contamination to NC and CC receptor regions, in particular to CC receptor region, as illustrated by its increasing ratios in Fig. 2.

Based on the modeled temporal trends of the contribution of the Southern China source to the soil burden in NE and CC from 1985, when the fresh use of the pesticide was ceased by regulation, to 2007, we can arrive at the following relationships for the change in contribution ratios in NE and CC receptor regions by fitting the modeled trends using a cubic linear equation:

For NE:

$$y = 7 \times 10^{-6}x^3 - 4 \times 10^{-4}x^2 + 0.0119x + 0.0636 \quad r^2 = 0.9995 \quad (2)$$

For CC:

$$y = 5 \times 10^{-6}x^3 - 3 \times 10^{-4}x^2 + 0.0083x + 0.0896 \quad r^2 = 0.9999 \quad (3)$$

where x is year and y is the contribution ratio.

Results from these two relationships indicate that contribution from the Southern China sources will increase to over 50% to the soil burden in the NE receptor region from 2030 and in the CC receptor region from 2043, respectively. This suggests that after these two years the Southern China sources will contribute more input of the chemical into these two receptor regions than the local sources. Overall, the analysis indicates that NE region is a major sink and the CC region is a weaker sink of the pesticide in China, even the Southern China source is closer to the NC than to the NE and CC regions.

Intensity of α -HCH use (Li et al., 1998b) and distance between source region and receptor region of a chemical (Li et al., 2010) strongly affect its soil residual levels in the receptor region. To further investigate the source–sink relationships, we have also estimated pollution equivalent (PE) that link the emission sources in Southern China and the soil burdens in the NE and NC regions, as well as the emission source in Central China and the soil residue in the NE region. The PE is defined as the ratio between the soil residual mass in the receptor regions and the accumulated usage in the source regions over the period from 1952 to the year of interested. Results

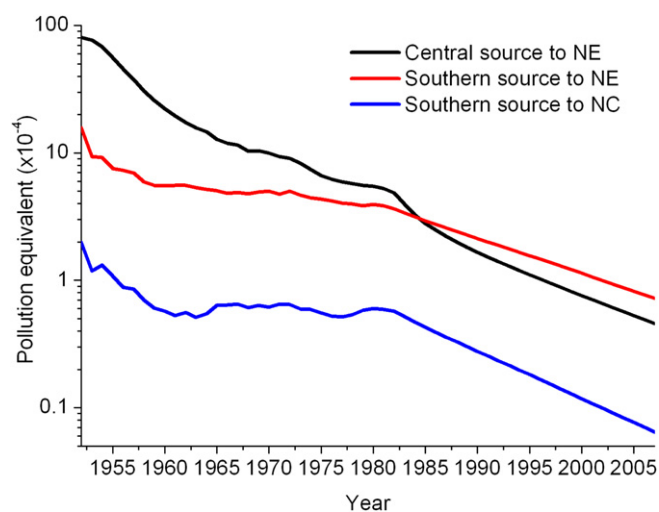


Fig. 3. Pollution equivalents of soil burden in NE receptor contributed by Southern source and Central source and in NC receptor contributed by Southern source.

are displayed in Fig. 3. The PE for the Southern China sources and the NE receptor region is about one order of magnitude greater than that for the NC receptor region over whole model integration period. This further demonstrates that the NE is the major sink region of α -HCH in China as compared with the NC region. For the period when α -HCH was still in use, the PEs related to the Central China source region were larger than that of the Southern China source region because of the fresh use of the pesticide and shorter distance between the sources and the NE receptor region. All the estimated PEs decrease with time. After 1984, the year when the pesticide was banned, the PE subject to the Southern China source has exceeded the PE value subject to the Central China source. In addition, the contributions of Southern China source to the NE receptor region are still higher than that to the NC receptor region as seen in Figs. 2 and 3, indicating that α -HCH from Southern China source is delivered to Northeastern China under favorable atmospheric circulation conditions for the northward atmospheric transport, as reported by Tian et al. (2009b).

5. Conclusion

Using the ChnGPERM, extensive modeling investigations have been carried out to assess α -HCH fate in Chinese environment from 1952 to 2007, and to detect sink regions in Chinese soil associated with environmental factors. We show that, although most of Eastern China soils were contaminated by α -HCH due to its heavy application in the past, Northeastern China has become a major sink of the pesticide. Given that most of POPs in China was applied in or emitted from Eastern China, such as DDT (Li et al., 1998a), PCBs (Xing et al., 2005), and PAHs (Zhang et al., 2007), the environmental behaviors of those chemicals having similar physicochemical properties as α -HCH were likely also driven by similar environmental factors. It is therefore deducible that the ambient temperature, SOC, atmospheric transport and deposition are important environmental factors and major pathways delivering and accumulating toxic chemicals from the south to north in the eastern part of China. Hence, for those toxic chemicals without fresh (primary) emissions, Northeastern China is very likely their major sinks driving by these pathways.

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Appendix. Supporting information

Supplementary information associated with this article can be found, in the online version, at doi:10.1016/j.envpol.2011.04.031.

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